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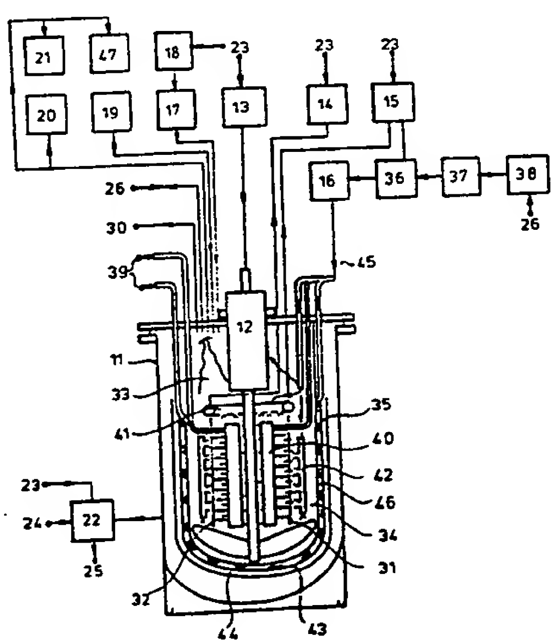
(54) **A process and apparatus for controlling the temperature of chemical reactions using liquefied gas as a coolant**

(57) The materials participating in the reaction are fed into a heat-insulating reactor 11 and are there cooled by means of a coolant at a lower temperature than the desired reaction temperature. Liquefied gas is used as coolant and is evaporated in the reactor 11, and the amount of liquefied gas charged-in is such as to ensure that in the reactor liquid, vapour and gas phases are all simultaneously present and the temperature of the discharging gas phase is maintained at a value lower than the desired reaction temperature. The means for passing coolant into the cooling system is an evaporator for evaporating liquefied gas, e.g. a coiled tube and/or pocket-shaped cooling insert(s) 40 and/or atomizing element(s) 41, 42 connected, expediently via a cryogenic valve 36, with a constant-pressure container 37 storing the liquefied gas. The interior 33 of the reactor 11 is expediently bounded by an exchangeable liquid vessel having a thin heat transfer surface 46.



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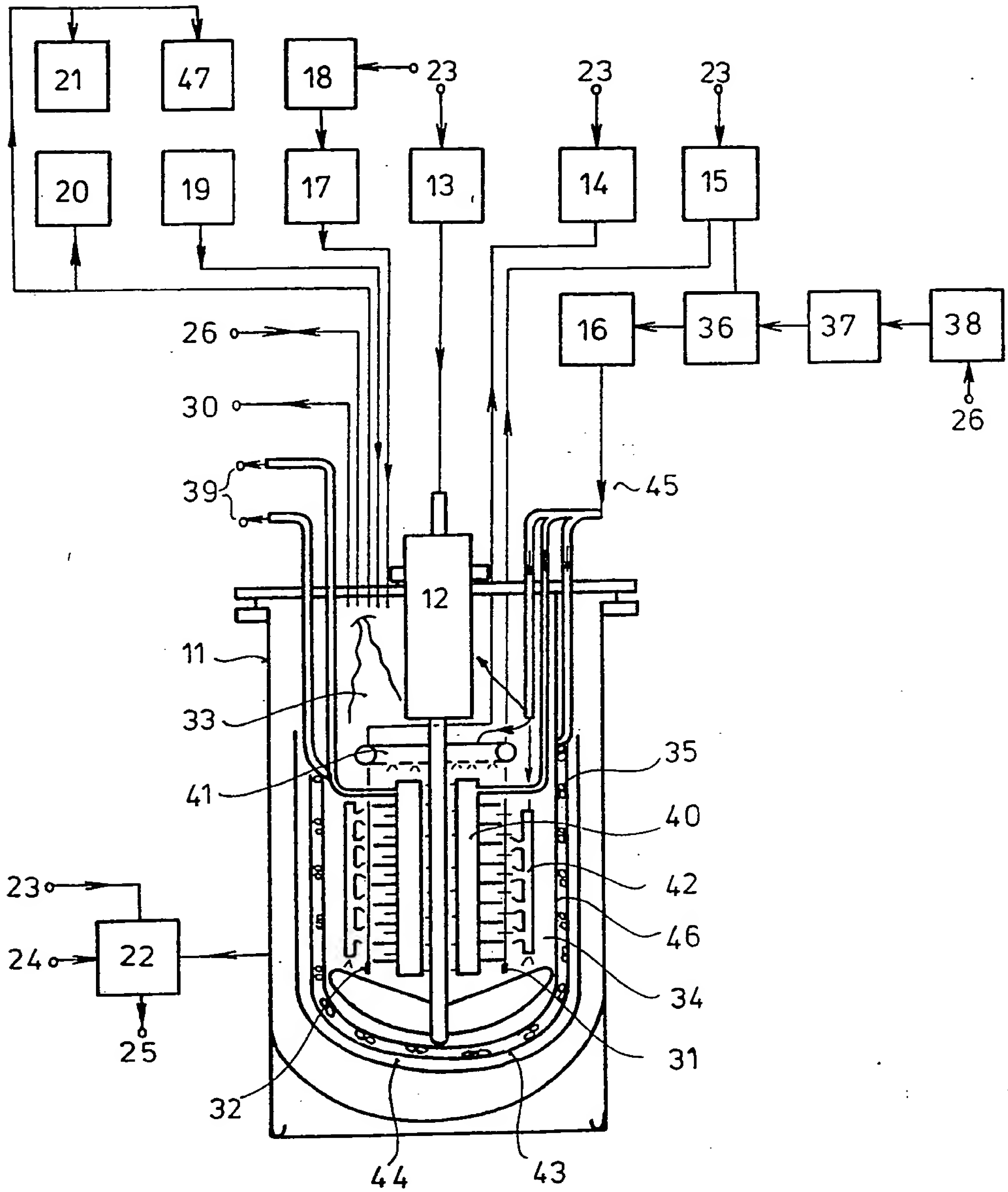
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A process and apparatus for controlling the temperature of chemical reactions

The invention concerns a process and apparatus for controlling the temperature of chemical reactions.

It is known that in certain chemical reactions a significantly increased quantity of final product of enhanced purity may be obtained from a given quantity of starting material by reducing the temperature of the reaction. This possibility is of primary significance particularly in the production of materials of high value and in cases where special requirements are prescribed with regard to the purity of the final product.

In the known solutions the materials participating in the reaction are fed to a heat-insulated reactor and are there cooled by means of a coolant of the desired reaction temperature, primarily by means of liquids of low freezing point and high specific heat. The heated-up coolant discharged from the reactor is pressed by a pump into a cooling apparatus and, after cooling to a prescribed extent, is recycled to the reactor.

In these known solutions it is a disadvantage that the cooling output is highly limited and may therefore only be used for small or medium quantities of the material and low reaction heat (see Gorenflo: Apparate fur die Kaltetechnik, [Equipments for Cooling Technology] Chem.-Ing.-Tech. 60 (1988), No. 11, C 855-858). It is also a disadvantage that the rate of cooling is relatively slow and requires a high quantity of coolant (cooling energy).

In the known apparatuses a coolant pump, cooling device and thermostat must be employed together. A further disadvantage is that the regulated region is of large dimensions and that the temperature regulator receives an error signal from the temperature sensor of the thermostat which has a relatively high time constant; consequently, even by adjusting the basic signal of the temperature sensor by means of the control temperature sensor in the reactor, cannot be regulated as regards the magnitude of the reaction temperature and the homogeneity of its

distribution within the optimum reaction time with the desired accuracy.

The aim of the invention is the obviation of the above drawbacks.

The invention aims to provide a process and apparatus for regulating the temperature of chemical reactions which enable an effective energy-sparing and accurate cooling or control even for high quantities of material producing high reaction heat.

The solution of the first task is a process for regulting the temperature of chemical reactions wherein the materials participating in the reaction are fed into a heat-insulated reactor and are there cooled by means of a coolant of lower temperature than the desired reaction temperature, and wherein according to the invention liquefied gas is employed as the coolant which is evaporated in the reactor, and the liquefied gas is introduced in an amount such that in the reactor, the liquid, vapour and gas phases are all simultaneously present, and the temperature of he discharged gas phase is maintained at a value lower than the desired temperature of the reaction.

Expediently, the liquefied gas is passed into the reactor intermittently.

Advantageously, the feeding of the liquefied gas is regulated as a function of the reaction temperature.

Expediently, the reaction temperature is sensed directly inside the material that is charged in.

Preferably, the liquefied gas is passed into the reactor from a container at constant pressure.

It is furthermore advantageous to atomize the liquefied gas in the reactor.

The atomization is performed either above the charged-in materials or within the charged-in materials themselves.

The solution of the second task is an apparatus for regulating the temperature of chemical reactions, comprising a heat-insulated reactor, a feeding device for charging the materials participating in the reaction into the reactor, a cooling system, means for passing coolant into the cooling

system, means for sensing the reaction temperature and a control unit which is connected with the means for sensing the reaction temperature and with the means for passing coolant into the cooling system, and wherein according to the invention the means for passing coolant into the cooling system is a means for evaporating liquefied gas, especially a coiled tube and/or pocket-like cooling insert(s) and/or atomizing element(s).

In a preferred embodiment of the apparatus according to the invention the means for evaporating liquefied gas is connected, expediently via a cryogenic valve, with a container storing liquefied gas at constant presure. According to another preferred embodiment, the container is connected via a reducer with the coolant outlet of the reactor.

Advantageously, the or each atomizing element is disposed above the space containing the materials participating in the reaction or is disposed in those materials themselves.

Expediently, the reactor is provided with a superinsulating covering.

The evaporating means, particularly the coiled tube is advantageously provided with a heat-conducting covering which is expediently formed from polished sheet aluminium.

Expediently, the evaporating means is a tube coiled in a bifilar configuration.

The constant pressure container is expediently connected with the output of the gas-liquefying apparatus.

The invention is described below by way of a preferred embodiment illustrated in the accompanying drawing which is a schematic diagram of preferred apparatus according to the invention.

The illustrated apparatus is connected to a reactor 11 equipped with an agitating device 12 coupled to a drive mechanism 13. The reactor 11 is provided with a diagrammatically illustrated inspection window 17 and illumination means 18. The materials participating in the reaction are passed into the reactor 11 by way of a feeder 19.

A control temperature sensor 31 immersed in the materials

charged in and a measuring sensor 32 are both arranged in the reaction space of the reactor 11. The temperature sensor 31 for control is connected with a control unit 15 while the temperature sensor 32 for measurement is connected with a temperature recording/display device 14.

Pocket-shaped cooling inserts 40 and atomizing elements 41, 42 are disposed in the interior 33 of the reactor 11. The atomizing element 41 is disposed above the materials 34 participating in the reaction while the atomizing element 42 is immersed in these materials. A bifilarly coiled tube 35 functioning as an evaporator is arranged in the boundary wall of the interior 33. The internal side of the wall is provided with a thin heat-transfer surface 46 while its outer side has a heat-conducting covering 43 made expediently from polished sheet aluminium. The covering 43 is surrounded by a superinsulating covering 44 of sandwich structure.

The coiled tube 35, the cooling inserts 40 and the atomizing elements 41, 42 are connected via a heat-insulated ducting system 16 and a cryogenic valve 36 to a container 37 maintained under constant pressure and containing a liquefied gas coolant 45, expediently liquid nitrogen. The coiled tube 35 and the cooling inserts 40 are provided with outlets 39 while the interior 33 has a coupling 26 connected with the container 37 by way of a stabilised reducer 38.

In the drawing, 30 designates a coarse (preliminary) vacuum system and 47 designates a rupturable safety diaphragm.

The reactor 11 has a safety valve 20 and a manometer 21. The illumination means 18, the temperature-recording unit 14 and the control unit 15 are connected to an electric power source 23. The reactor 11 is additionally provided with a vacuum system 22 which is also connected with the electric power source 23 and has an inlet 24 and an outlet 25 for cooling water.

In the apparatus described above, the process according to the invention takes place as follows:

The materials 34 participating in the reaction are passed into the interior 33 of the reactor 11 by means of the feeder 19

and are mixed with the aid of the agitating device 12. By intermittently opening the cryogenic valve 36 liquid nitrogen coolant 45 is passed from the container 37 through the ducting system 16 into the coiled tube 35 wherein it is evaporated. The evaporating coolant 45 intensively cools the materials 34 and is then discharged via outlet 39 in the form of a gas of a temperature lower than the desired reaction temperature. The liquid nitrogen coolant 45 is similarly passed into the cooling inserts 40 wherein it is again evaporated and discharged in gaseous form through the outlets 39 of the coolant inserts. With the aid of the atomizing elements 41 and 42 the coolant 45 can be evaporated in an atomized form above the materials 34 as well as within the materials 34. In this way, extraordinarily intensive <u>in situ</u> cooling may be achieved.

The opening and closing of the cryogenic valve 36 is controlled by the control unit 15 on the basis of the signal of the temperature sensor 31 and in this way the inertia of the system is low, i.e. the risk of the reaction 'running away' or 'choking' is low. Naturally the rapid reaction is also promoted by the fact that not only the specific heat but also the heat of the evaporation are used for cooling.

By way of the coupling 26 connected with the interior 33 an inert protective gas atmosphere is formed above the materials 34 by means of dry nitrogen as at reduced pressure. With the aid of the vacuum system 22 a desired amount of evacuation may be assured in the reactor 11 in order to maintain heat insulation.

Naturally it is possible to operate all, or only a part, of the coiled tube 35, the coolant inserts 40 and the atomizing elements 41, 42 in accordance with the actually required cooling output. The control unit 15, which is expediently a two-position regulator of PD characteristic, so controls the operation of the cryogenic valve 36 (e.g. electromagnetic valve) that the discharging coolant 45 should always be in its gaseous phase.

The coiled tube 35 and the cooling inserts 40 (or other evaporating means) may be formed as countercurrent or cross-current heat-exchangers. For instance, it may be expedient to use

finned heat-exchanges or hot foil-type heat-exchangers. In given cases the container 37 may be connected to the output of a gas-liquefying apparatus, but is may also be an exchangeable transport (delivery) vessel.

In a suitable case the inner vessel of the reactor 11 itself may be exchangeable in order to match the actually required volume.

The use of the process and apparatus according to the invention has the following advantages:

- The evaporative enthalpy of the liquefied gas and the enthalpy gained by the heating up of the cold gas are utilised directly for cooling the charge in the reactor.
- Because of the high heat transfer coefficient characteristic of a phase change the heat transfer between the coolant and the charge is very good.
- The cooling process takes place in a well-insulated space and directly next to and/or in the charge and thus losses of cooling energy are very small.
- Even for large charges the cooling is rapid and good temperature control may be achieved.
- The cooling range may be extended to $-170^{\circ}C$.
- Because of the heat transfer between the inlet and outlet channels of the coolant a homogeneous temperature distribution may be assured.
- The reaction temperature may be regulated with high precision.
- In the case of small charges and small heat loads dry, cold gaseous coolant (nitrogen gas) may also be used.
- The appropriate reaction time may be reliably set even for higher heat loads.
- Even in the event of manually feeding the materials participating in the reaction the desired reaction temperature may be stabilized with the appropriate accuracy; alternatively, the feeding may be simply automated at little extra expense.
- The apparatus according to the invention is of simple

construction, operationally reliable, does not involve high investment and operating costs and its servicing/maintenance is simple.

- When one utilizes exchangable reaction vessels of different charge capacities the apparatus becomes suitable both for carrying out laboratory experiments and for experimental manufacture.
- The apparatus is widely utilizable, i.e. is versatile.
- The final product yield is very favourable.
- The final product is purer than is usual.

In the annexed claims reference numbers have been used purely by way of example in order to facilitate comprehension, but it is hereby declared that absolutely no limitation of scope whatsoever is intended thereby.

CLAIMS

1. A process for controlling the temperature of chemical reactions, comprising feeding the materials participating in the reaction into a heat-insulated reactor and cooling them there by means of a coolant having a temperature lower than the desired reaction temperature, wherein as coolant liquefied gas is used which is evaporated in the reactor and the liquefied gas is fed into the reactor in an amount such that liquid, vapour, and gas phases are all simultaneously present in the reactor, and the temperature of the discharged gas phase is maintained at a value lower than the desired reaction temperature.

2. A process according to claim 1, wherein the liquefied gas is introduced into the reactor intermittently.

3. A process according to claim 1 or 2, wherein the feed rate of the liquefied gas is regulated as a function of the reaction temperature.

4. A process according to claim 3, wherein the temperature of the reaction is sensed directly within the charged-in materials.

5. A process according to any preceding claim, wherein the liquefied gas is passed into the reactor from a container maintained at constant pressure.

6. A process according to any preceding claim, wherein the liquefied gas is atomized in the reactor.

7. A process according to claim 6, wherein the atomization is performed above the charged-in materials.

8. A process according to clim 6, wherein the atomization is performed within the charged-in materials.

9. Apparatus for controlling the temperature of chemical reactions, comprising a heat-insulated reactor, a feeder for passing materials participating in the reaction into the reactor, a cooling system, means for passing coolant into the cooling system, and means for sensing the reaction temperature and a control unit connected with the means for sensing the reaction temperature and with the means for passing coolant into the

cooling system, and wherein the means for passing coolant into the cooling system is an evaporating means for evaporating liquefied gas.

10. Apparatus according to claim 9, wherein said evaporating means is a coiled tube (35) and/or pocket-shaped cooling insert(s) (40) and/or atomizing element(s) (41, 42).

11. Apparatus according toclaim 9 or claim 10, wherein the means for evaporating liquefied gas is connected with a container (37) maintained at constant pressure storing the liquefied gas.

12. Apparatus according to clim 11, wherein said evaporating means with said container (37) by way of a cryogenic valve (36).

13. Apparatus according to laim 11 or 12, wherein the container (37) is connected via a reducer (38) with at least one coolant discharge outlet (26) of the reactor (11).

14. Apparatus according to any of claims 9 to 13, wherein an atomizing element (41) is arranged in the space above the materials participating in the reaction.

15. Apparatus according to any of claims 9 to 13, wherein it has an atomizing element (42) arranged within the materials participating in the reaction.

16. Apparatus according to any of claims 9 to 15, wherein the reactor (11) has a superinsulating covering (44).

17. Apparatus according to any of claims 9 to 16, wherein the evaporating means is provided with a heat-conducting covering (43).

18. Apparatus according to claim 17, wherein said covering (43) is formed from polished sheet aluminium.

19. Apparatus according to any of claims 9 to 18, wherein the evaporating means is a bifilarly coiled tube coil (35).

20. Apparatus according to claim 11 or any claim appendant thereto, wherein the constant pressure vessel (37) is connected with the output of a gas-liquefying apparatus.

21. Apparatus according to any of claims 9 to 20, wherein the interior (33) of the reactor (11) is bounded by an exchangeable liquid vessel having a thin heat transfer surface

(46).

22. A process according to claim 1, substantially as herein described with reference to and as shown in the accompanying drawing.

23. Apparatus according to claim 9, substantially as herein described with reference to and as shown in the accompanying drawing.

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